Deliquescence Of Fission Product Salts: Implications for the Corrosion Behavior of Spent Fuel

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Introduction

- Our work focuses on strengthening the scientific basis for safely storing and disposing of spent nuclear fuel from commercial power plants.
- We are performing experiments to quantify processes that may initiate the aqueous corrosion of spent fuel exposed to humid air. These processes may lead to the release of radionuclides into the environment.
- Our results will facilitate the development of mechanistically based predictive models that are needed to assess the safety performance of nuclear waste repositories.

Challenges

- Spent fuel will be the hottest (thermally) material in a repository due to radioactive decay (Figure 1). The temperature difference between the fuel and tunnel wall results in a water vapor pressure gradient that drives water vapor away from the spent fuel surface.
- However, deliquescent minerals in the spent fuel may lower the water vapor pressure at the spent fuel surface enough to cause water condensation despite the thermal gradient (Figure 2).
- For our research to be successful we must quantify the deliquescence behavior (i.e. relative humidity threshold for water condensation) of any hygroscopic mineral phases that may be present in spent fuel (under relevant thermal and humidity conditions).

Our Approach

- We are using an isopiestic approach to study the deliquescence behavior of hygroscopic minerals that may be present in spent fuel (Figure 3).
- Reaction products from the isopiestic experiments will be characterized using X-ray Absorption Spectroscopy, X-ray diffraction, Electron Microscopy, and Inductively Coupled Plasma Mass Spectrometry.

Physical/Chemical Scenario Under Investigation

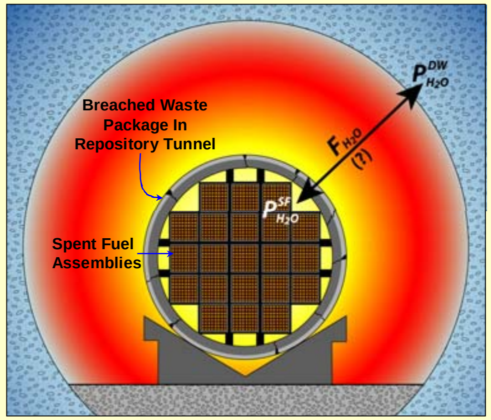

Figure 1: Cross-section through a geologic repository tunnel showing a breached waste package containing commercial spent nuclear fuel. We are studying how the presence of humid air in such a scenario could initiate the degradation of the spent fuel. Deliquescent fission product minerals in the fuel could lower the water vapor pressure (P^{SF}) at the spent fuel surface causing a flux of water (F_{H2O}) toward damaged fuel pins (P^{DW} = vapor pressure at the tunnel wall).

Process Under Investigation

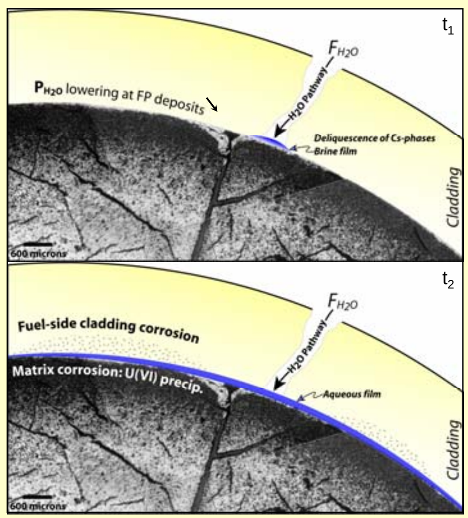

Figure 2: Reflected light micrograph of a cross-section through a spent fuel pellet. These images illustrate how deliquescence could initiate the aqueous corrosion of spent fuel exposed to humid air (FP = fission products).

Adapted from material provided by Hanchung Tsai, Argonne National Laboratory (ANL), Energy Technology Division, NRC Review of ANL High-Burnup Cladding Performance Program, 2003.

Experimental Approach

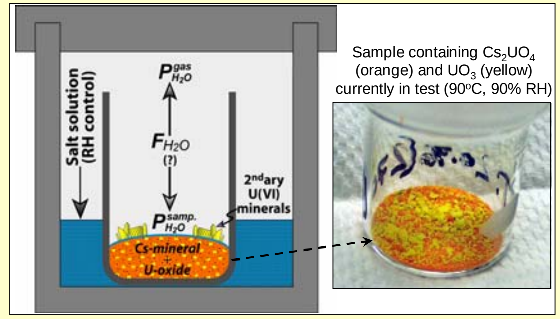

Figure 3: Schematic diagram of the experimental vessel used to quantify the deliquescence behavior of fission product salts. This setup employs an isopiestic approach in which a salt solution is used to control the relative humidity (water vapor pressure) surrounding the sample. These experiments are performed in calibrated ovens over a range of 40 – 90°C.

Results Summary

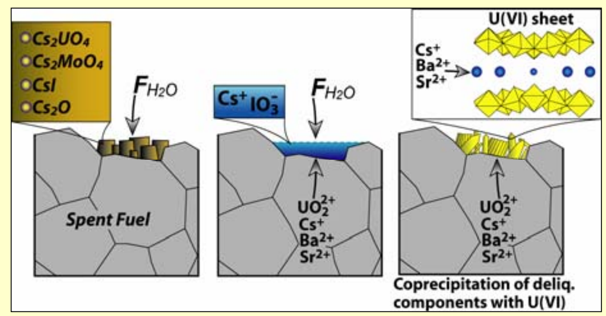

Figure 4: Results to date indicate that if spent fuel containing cesium-bearing fission products is exposed to air with a relative humidity >80% water may condense on the fuel even if the fuel is ~5°C hotter than its surrounding environment. Water will accumulate at the fuel surface due to water vapor pressure lowering caused by deliquescent components (e.g., Cs, Ba, Sr). However, the sequestration of these deliquescent components in U(VI) corrosion phases that form as the fuel degrades will counteract vapor pressure lowering and water accumulation. This sequestration process will thus promote evaporation and therefore limit the amount of water (e.g., brine film thickness) that accumulates at the spent fuel surface.

Results

- Our findings (Figure 4) indicate that deliquescence will be an important process by which water first contacts spent fuel exposed to humid air in a geologic repository. However, the process will most likely be self-limiting due to the sequestration of deliquescent components in spent fuel alteration phases.

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